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A modelling approach for estimating background pollutant concentrations in urban areas

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ABSTRACT

Air pollution control strategies have helped to improve the air quality over Europe over the last twenty years. Despite this success, the improvements have been insufficient to protect health of those who spend most of their time within urban areas and particularly near major roads. Given the inherent complexity of urban environments and the incomplete understanding of the physical and chemical processes involved in pollutant dispersion, it is a challenging task to estimate urban air quality. In order to address this issue, a new regression model for estimating the urban increment for all cities of a region up to Europe as a whole is developed. The model is able to capture the higher pollutant concentrations commonly found within urban areas for assessing the localized effects associated to urban emissions. This approach is used for estimating annual concentrations of PM_{10} and NO_2 for all urban areas with more than 50 000 inhabitants in Germany for the reference year 2005. The results showed that there are differences on air quality levels across urban areas in Germany. This information is relevant when evaluating the impact of emission reduction policies on air quality, which should take into account the cost–benefit of each measure. Furthermore, as the modelling approach allows for flexibility changing initial conditions and building scenarios, the added values of this approach are the large spatial domain covered, the high spatial resolution, and its inherent flexibility to address other environmental issues, such as analysis of emission reduction scenarios, the human exposure to certain pollutants and their associated human health impacts.

Keywords: *Urban air quality, urban increment, regression model, PM_{10}, NO_X*

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1. Introduction

Air pollution control strategies have helped to improve the air quality over Europe over the last twenty years (EEA, 2010). However, the concentration limits of the Air Quality Directive of the European Union for PM_{10} and NO_2 (CEU, 1999) are frequently exceeded in European cities (Jenkin, 2004a; Chaloulakou et al., 2008). Even more alarming is, that the estimated health impacts due to high $PM_{2.5}$ concentrations lead to a considerable reduction of the life expectancy of the European population and to other health risks (Amann et al., 2005). Over 75% of the European population is living in urban areas and by 2020, this will be 80%, and for some countries even 90% (EEA, 2006). As a consequence, the number of people affected by elevated concentrations of pollutants is notably higher in urban areas than in rural environments. Trying to shed light on the mechanisms accountable for the typical urban pollution increment for most pollutants, the topic has become the subject of a number of studies over the last few decades (De Leeuw et al., 2002; Yin et al., 2005; Charron et al., 2007; Chaloulakou et al., 2008; Stedman and Derwent, 2008).

In this context, further measures to reduce PM_{10}, $PM_{2.5}$ and NO_2 concentrations in European cities are necessary. However these measures generally cause costs and thus have negative economic impacts. It is thus essential and mandatory to prove with a cost benefit analysis that the measure is not only effective, i.e. leads to the desired reduction in urban concentration, but efficient as well, i.e. showing that the avoided damages due to reduced concentration are higher than the costs. An example of this practice is the cost–benefit analysis of the Clean Air for Europe (CAFE) programme leading to the new air quality directive (Holland et al., 2005a).

Physical impacts attributed to a certain activity are estimated using Concentration Response Functions (CRF) (Holland et al., 2005b; Hurley et al., 2005). Using CRFs it is possible to estimate the health risks caused or avoided due to changes in the annual average urban background concentrations (NEEDS, 2007). In addition, thresholds for the $PM_{2.5}$ annual average urban background concentrations are given in the Air Quality Directive (CEU, 2008). It follows that for preparing and assessing air pollution control policies and measures, the urban background concentration of pollutants is needed; and that not only for one city, but – as national and EU–wide policies and measures need to be analysed – for all cities in a country or the whole EU. The tools to estimate concentrations for emission scenarios are atmospheric models. And indeed, several atmospheric dispersion models have been developed over the last decades to estimate pollutant concentrations for different spatial scales, such as regional scale (Ebel et al., 1997; Sartelet et al., 2007; Schaap et al., 2009), urban scale (De Leeuw et al., 2001; Mediavilla–Sahagun and ApSimon, 2003; Mensink and Cosemans, 2008), and even at street level (Huang et al., 2009; Jenkin, 2004b; Murena et al., 2009). For estimating the concentrations in a particular city, nesting approaches have been developed to handle multi–scale problems covering from regional to local areas and simulate specific areas with a higher spatial resolution (e.g., Brucher et al., 2000; Kessler et al., 2001; Tang, 2002). However, the applicability of this approach is limited to one or a few urban areas, as requirements for input data and computational resources increase strongly with increasing size of

the inner domain. As a nesting approach for all cities of a larger region or Europe is not yet possible, other combined approaches have to be developed in order to take advantage of specific model strengths. Such approaches have been proposed e.g. by Gokhale and Khare (2005) (deterministic model with statistical components), Sahlodin et al. (2007) (Gaussian model with a computational fluid dynamics model) and Mensink and Cosemans (2008) (Gaussian model for street canyons). Despite the good results obtained by these approaches, they cannot be applied for the purposes followed in this work: to provide estimates not only for specific cities but for all major cities in a large domain, such as Germany. Moreover, flexibility of changing initial conditions and building scenarios is another issue to be addressed.

We propose to combine the results of two models: first, a regional atmospheric CTM (chemistry–transport–model) that provides pollutant concentrations at a regional scale, which are usually calculated for large grid cells containing both rural and urban areas; a second simpler model that provides the difference between the background concentration in a city and the concentration outside of the city. In this way, it is possible to estimate the annual average urban background concentration for all cities within a large region by adding both concentrations.

Some attempts have already been made for estimating this urban increment (i.e., the difference between the average background concentration in a city and the concentration measured outside the urban area) for large cities in a generalised way Europe–wide, especially by De Leeuw et al. (2001) and the CityDelta project (Cuvelier et al., 2007; Thunis et al., 2007). Even though the CityDelta project provides an important contribution to the understanding of the mechanism involved in the urban increment, some issues remain to be addressed. For instance, the characterisation of urban areas is mostly defined by their administrative boundaries or diameters. Yet, these parameters do not provide information concerning the actual extent of the urban sprawl. This distinction is vital for addressing the difference between pollution levels within urban areas with respect to rural levels and this fact may have influence on the estimation of the corresponding urban emissions. Furthermore, land use changes over time may not be adequately captured if the only parameter to characterize the size of a city is its diameter. Another issue to consider is that the variability in the urban increment across all countries within Europe can be expected to be large due to the heterogeneity in meteorological, topographical and emission release conditions. This suggests that a unique functional relationship for all European countries may not be able to capture such differences across countries. In addition, the quality of the emission data used is of course important for the quality of the results.

Addressing the issues pointed above, a new regression model for estimating the urban increment for all cities of a region up to whole Europe is developed. The approach is tested and used for all major cities in Germany. The model is able to capture the higher pollutant concentrations commonly found within urban areas for assessing the localized effects associated to urban emissions. The modelling approach is used for estimating the urban increment for annual mean PM_{10}, $PM_{2.5}$, and NO_2 concentrations for all urban areas with more than 50 000 inhabitants in Germany for the reference year 2005. Furthermore, a comparison against measured values is also presented.

2. Methodology

In order to gain insight into air quality trends in Germany, measured data for all German cities were retrieved from the European Air Quality database Airbase (Airbase, 2010). In Figure 1, annual mean PM_{10} concentrations for rural and urban background, along with traffic urban monitoring station types in Germany are depicted. According to the EuroAirnet (European Air Quality Monitoring Network) criteria, rural background stations are used for monitoring the air pollution levels resulting from long–range transport of air pollutants and from emissions in the region in which the station is located. These stations are commonly located 10–50 km from large pollution sources, such as cities, power plants and major roads. On the other hand, urban background stations (including suburban background stations) measure the average air quality in urban areas, resulting from the regional concentration and from emissions generated within the city, not reflecting the direct influences by large emission sources (i.e., industrial plants or busy streets) (Larssen et al., 1999).

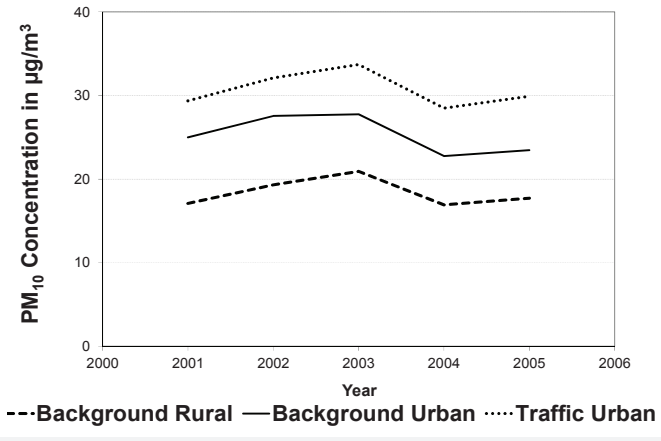

***Figure 1**. Changes in PM_{10} annual mean concentrations in Germany based entities over the period of 2001-2005. Source: AirBase (2010).*

Pollutant concentrations significantly increase as one moves from regional into urban and further into traffic–influenced areas (street canyons) (Figure 1). Indeed, it should be evident that pollutant concentrations are generally higher with increasing proximity to the corresponding emission sources, in this case PM_{10}. It is worth noting here that similar trends for PM_{10} and NO_2 as in Germany can be observed across Europe (Mol et al., 2008).

Thus, considering how pollutant concentrations vary across monitoring stations, it is reasonable to argue that the total pollutant concentrations measured at urban background sites can be considered as being composed by two distinct concentration layers: the regional layer, comprising the rural background concentration; and the urban layer, depicting the concentration increment normally found within cities. It can be furthermore suggested that each layer can be estimated individually, using the better approach for the corresponding scale and building on the subjacent layer. This concept is the basic idea behind the modelling approach (i.e., regional background concentrations plus the urban increment model) presented in the present study.

Here it is important to remark that not all pollutants behave in the same way as yearly average PM_{10} or NO_2 concentrations. In fact, when calculating the same averages for ozone the trend may be reversed (e.g. Munir et. al., 2012). One reason for this is that road transport contributes to ozone depletion near roads through the so–called titration process. This process consists of the removal of ozone through reaction with nitric oxide in the vicinity of NO_X emission sources (Pison and Menut, 2004; Liu et al., 2007). Furthermore, ozone is a highly reactive oxidant that interacts with a number of other compounds, the rates of which also depend on location variables, such as air temperature and meteorological conditions.

2.1. Model structure

In an effort to provide a robust air quality information with less demanding atmospheric dispersion modelling, several

attempts have been made for estimating the urban increment for large cities, for instance by De Leeuw et al. (2001) and, more recently, by the CityDelta project (Cuvelier et al., 2007; Thunis et al., 2007). They have attempted to calculate Europe–wide urban concentrations in a generalised way with low computing time and input data by means of parameterising the most important phenomena. An improved and extended approach was followed in the present work, assuming that the urban pollution increment can be explained in terms of the emissions released within the city, urban morphology and average wind speed. In the following, the components of the urban increment model are described.

Measured urban increment. An urban increment is defined here as the distinguishable difference between the background concentration in a city and the concentration measured outside the urban area. The urban pollution increment for several urban areas was estimated using observational data from paired rural and urban background measuring stations. For this purpose, measured annual mean PM_{10} and NO_X concentrations for urban background (including suburban background) (UBS) and rural background stations (RBS) were retrieved from the European Air Quality database (AirBase). Concerning $PM_{2.5}$, the limited availability of measurements hampered developing the estimation of the measured urban increment for this pollutant. An alternative approach was used for making a rough estimation of $PM_{2.5}$ urban increment, namely the $PM_{2.5}/PM_{10}$ concentration ratio, which is discussed later in this paper.

Although NO_2 (not NO_X) concentrations are needed, the regression analysis is made for NO_X, to avoid considering the chemical transformation between NO and NO_2 in the analysis. The derived NO_X concentration increments are then converted into NO_2 concentration increments using an empirical function which is introduced in Section 2.5.

Annual mean concentrations based on hourly values for urban and rural air quality stations with over 80% valid measurements were included in this analysis. Using spatial analysis, urban background stations located within urban areas with more than 50 000 inhabitants were considered. When several UBS were found within the same urban entity, an average urban background concentration for each urban entity was estimated using all UBS of the city. The main reason for this simplification lies in the fact that, as the urban entity is viewed as the unit of analysis, only one urban increment for each urban entity was considered to be enough for characterizing such unity. Further disaggregation within the urban entity was tested. However, the gains in accuracy did not outdo the larger uncertainties resulting from using coarse wind fields and the lack of specific urban morphology data.

Each UBS was associated with their nearest RBS and, restricting the distance between them to 10–50 km, pairs of stations for PM_{10} and NO_X were built. Ideally, a RBS should be chosen in such a way that it is located upwind with respect to the UBS in order to avoid a significant influence of urban emissions on the measurements at the rural station. However, wind direction varies on a daily and seasonal basis. Furthermore, it is possible that for a given location there is more than one dominant wind direction or a wide range of dominant winds, requiring the use of several rural stations for estimating the difference between urban and rural pollution levels (e.g., Jones et al., 2008). Given that such detailed analysis goes beyond the scope of this work, this aspect was not investigated. Nevertheless, it is important to note that, by definition, rural stations are located within a distance of 10 to 50 kilometres from built up areas and other major sources. Despite this distance, measurements from rural stations may be partially influenced by regional traffic activity, larger power plants, and industrial complexes. Therefore, a certain influence of urban emissions on pollution levels measured at rural stations is likely unavoidable.

A total of 24 pairs of stations for PM_{10} and 26 pairs of stations for NO_X were identified and the urban increment were then computed as the difference between the corresponding urban/rural stations. Half of the station pairs were used for model development and the other half was used for model evaluation, which is discussed in Section 4. The PM_{10} urban increment values used for model development are summarised in Figure 2 and similar data were available for NO_X (see Figure S1 in the Supporting Material, SM).

The urban increments estimated above constitute the dependent variable to be used in the multiple regression analysis presented later in this Section.

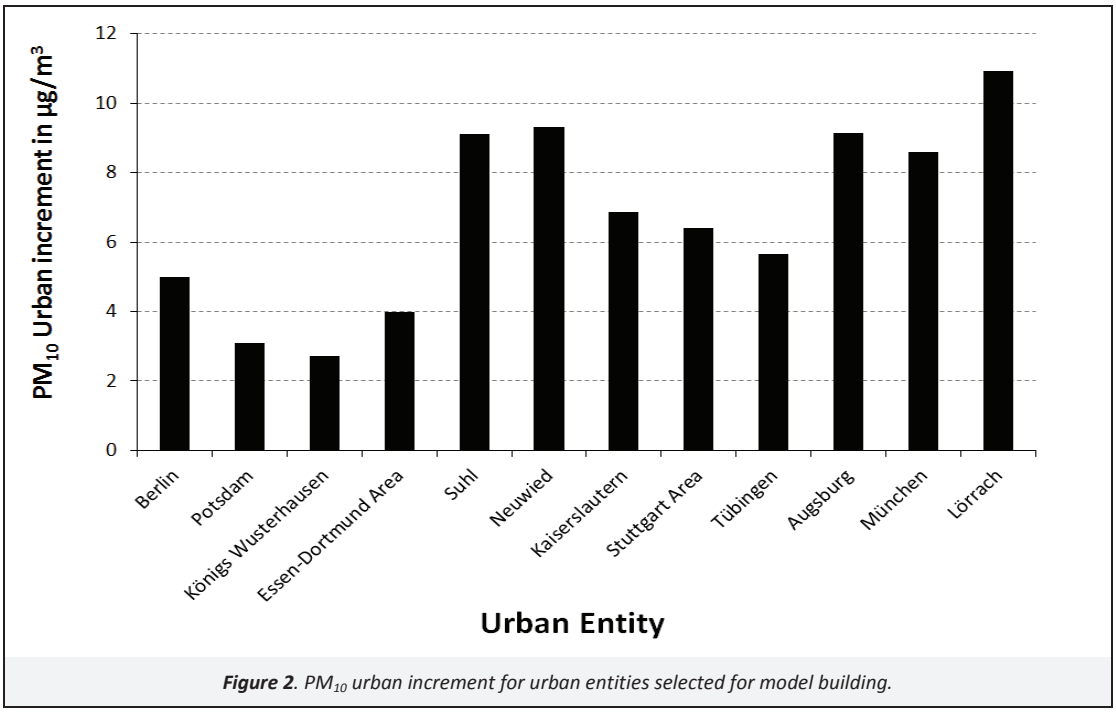

***Figure 2**. PM_{10} urban increment for urban entities selected for model building.*

Urban emissions. Highly spatially disaggregated urban emissions were provided by the Institute for Energy Economics and the Rational Use of Energy (IER), University of Stuttgart. Yearly emissions with a horizontal resolution of a 1x1 arc–minute for the year 2005 were used in this work and the main pollutants considered were primary PM_{10}, $PM_{2.5}$, and NO_X – all of them aggregated into eleven SNAP (Selected Nomenclature for Air Pollution) categories at the level one. This emission dataset has been developed for and successfully applied in several national and EU projects, such as GENEMIS, ESPREME, (Friedrich et al., 1999; Kuhlwein et al., 2002; Friedrich and Reis, 2004) and more recently, PAREST (Builtjes et al., 2008). Furthermore, it is also assumed that only primary emissions released from low–height sources increase concentrations of the respective pollutants within the cities (Amann et al., 2007); the provided emissions were classified according to their release heights following Pregger and Friedrich (2009).

Urban morphology. Relevant to any discussion involving urban areas is defining what areas are considered as such. Due to significant definition differences among countries, there is no international agreement upon the definition of rural and urban areas (UN, 2007). In recognition of the importance of defining urban boundaries when addressing urban pollution, the methodology for defining Urban Morphological Zones (UMZ) proposed by Milego (2007), which merges land–use classes contributing to the urban tissue and function, was adopted in this work. Assuming that additional pollution levels generated by smaller UMZ are low or negligible, only urban areas with a population larger than 50 000 inhabitants are analysed in more detail. Applying this criterion, there are 114 such entities in Germany comprising about one third of the total population.

Wind speed. A wind–speed dataset modelled within the EU project NATAIR (Vautard, 2006) was integrated into this analysis. Profiting from a four–year dataset (1997, 2000, 2001 and 2003) and a spatial resolution of 25x25 kilometres, representative values for average 10–metres–over–the–surface wind–speed was estimated for each urban area under study.

2.2. Multiple–regression analysis

After all parameters, namely emission intensity, average wind speed, and rural background concentration were tested for statistical significance at 0.05 level, a multiple–regression analysis was conducted to predict urban increment from the predictor variables discussed in this Section. The equation is based on previous work developed within the CityDelta project (Cuvelier et al., 2007; Thunis et al., 2007) with the improvements mentioned in the introduction. Thus, the urban increment was estimated using the following equation:

$$C_{i\,UrbInc} = \omega_i + \varphi_i \frac{E_{i\,UE}}{A_{UE} \cdot U_{avg}} + \gamma\, C_{i\,rural} \qquad (1)$$

where $C_{i\,UrbInc}$ is the urban increment of pollutant *i* in µg/m^3, $E_{i\,UE}$ is the total emission of pollutant *i* within the urban entity in tons, A_{UE} is the area of the urban entity in km^2, U_{avg} is the average wind speed in m/s, $C_{i\ Rural}$ is the rural background concentration of pollutant i in µg/m^3, ω_i, φ_i, and γ_i are the multiple–regression parameters for pollutant *i*, dimensionless.

The multiple–regression analysis results are presented in Table 1.

Using the model with the coefficients depicted in Table 1, it is possible to estimate the urban increment concentration for PM_{10} and NO_X. Furthermore, the adjusted R^2 gives information about the influence of additional variables in the model. It is included in the results as this metric was used in the process of model development.

Table 1. *Multiple-regression parameters and coefficients of determination*

Parameters	PM_{10}	NO_X
ω	15.27	-6.95
ϕ	0.24	5.64
γ	-0.53	n.s.
R^2	0.80	0.83
Adjusted R^2	0.75	0.82

n.s.: not significant

2.3. $PM_{2.5}$ urban increment

As already mentioned, an urban increment model for $PM_{2.5}$ could not be developed directly due to the limited availability of valid measurements for this pollutant. However, considering that ambient concentration levels of this pollutant are relevant to assessing human health, an alternative approach was used for making a rough estimation of $PM_{2.5}$ urban increment, namely the $PM_{2.5}/PM_{10}$ concentration ratio. A number of studies have been conducted to shed light on the ratio $PM_{2.5}/PM_{10}$. For instance, Gehrig and Buchmann (2003) analysed long–term data from several urban monitoring stations in Switzerland noting that the mean $PM_{2.5}/PM_{10}$ ratios of daily values were 0.75 with a standard deviation of 0.11. They also found that the ratio remained rather constant at the different stations and through a period of four years. Van Dingenen et al. (2004) carried out a comprehensive analysis of the physical characteristics of particulate matter using data from 31 monitoring stations across Europe. They report that the $PM_{2.5}/PM_{10}$ ratio vary between 0.57 and 0.85, depending strongly on the type of monitoring station. In the case of urban background stations, the ratio ranged between 0.66 and 0.80. Moreover, combining measurements and detailed dispersion modelling, Graff (2006) evaluated the $PM_{2.5}/PM_{10}$ ratio for seven large regions in Germany, finding that the proportion of $PM_{2.5}$ in PM_{10} urban background ranged from 0.70 to 0.75. Thus, although it is recognized that the ratio between $PM_{2.5}$ and PM_{10} depends on factors which vary both spatially and temporally, and that there is no single ratio value valid across all measurement stations (Putaud et al., 2010). Thus, a value of 0.75 for the $PM_{2.5}/PM_{10}$ urban background ratio is considered to be a reasonable assumption. The reason for choosing the upper limit lies in the fact that current models underestimate PM_{10} concentrations (Vautard et al., 2007) and this value might provide estimates closer to the actual concentrations. Nevertheless, due to the inherent spatial and temporal variation in $PM_{2.5}/PM_{10}$ ratio, it should be kept in mind that that this approach should only be used when no other data is available.

2.4. NO_X to NO_2 conversion

We have developed a model to estimate the urban increment concentration for NO_X. However what needed is the concentration for NO_2, as thresholds and concentration–response relationships referring to NO_2. A simple model is applied to derive NO_2 concentrations from modelled NO_X concentration values, namely the Romberg model (Romberg et al., 1996; Bachlin et al., 2006) which is based on a regression analysis of NO_2 and NO_X measurements from the German monitoring network. According to this model, annual average NO_2 concentrations at roadsides are computed from annual average NO_X concentrations as follows:

$$NO_2 = \frac{A\,[NO_x]}{([NO_x] + B)} + C\,[NO_x] \qquad (2)$$

where NO_2 and NO_X are the annual average concentrations of those pollutants; *A*, *B*, and *C* are the correlation constants with values 103, 130, and 0.005, respectively.

Here it is important to note that these coefficients are based on measurements made between 1987 and 1993 and, ever since, an increasing trend in the NO_2/NO_X emissions ratio from road traffic exhaust emissions has been observed (Scholz and Rabl, 2006). Therefore, the correlation constants presented above are not suited to describe current NO_2 concentrations at roadsides. In order to take into account this development, Bachlin et al. (2006) updated the Romberg function using measured data for the years 2000 to 2003. From this revision, the values for the correlation constants *A*, *B* and *C* from Equation (2) were updated as 43, 53, and 0.129, respectively.

Due to its ease of application and flexibility, this method has been used in several studies. For instance, Puxbaum et al. (2003) measured NO and NO_2 concentrations in the vicinity of a tunnel portal for almost a year. Satisfactory predictions of NO_2 using the Romberg Method suggest that its use may be regarded as a satisfactory alternative to more complex models. Empirically derived functions to determine the proportion of NO_X as NO_2 have been used in several studies. Derwent and Middleton (1996) developed an empirical function for the ratio of NO_2 to NO_X derived from monitored data at an urban road site in London. Owen et al. (2000) used this function along with a full chemical model for predicting NO_2 concentrations in a large urban area. The results showed that the empirical function provided better agreement with measurements than the complex model.

Thus, as the arguments presented above suggest, it seems reasonable to assume that the parameterised function to derive annual average NO_2 concentrations from modelled annual average NO_X concentration values is an acceptable alternative for more complex full chemical and dispersion models. Nevertheless, it is worth noting that an application for a more recent year (e.g., 2012) would certainly need a further update of the model's coefficients.

2.5. Coupling with regional chemistry–transport models

Chemistry–transport models are able to provide concentration estimates for large domains. They are a valuable tool for assessing regional trends over a certain time scale. However, current models have difficulty in capturing higher concentrations commonly found in urban areas for certain pollutants (Vautard et al., 2007). Therefore, the urban increment approach could be used to correct for this underestimation. However, if we assume that the concentrations derived by the regional model for a grid cell is correct, the urban increment cannot simply be added to the concentration in the grid cell. In this case, double–counting of emissions should be avoided adjusting the urban and non–urban concentrations in the grid cell in such a way, that the area–weighted average concentration stays the same as before. On the other hand, if regional background concentrations near urban areas are available, the urban increment concentration can be added directly.

3. Results and Discussion

The PM_{10}, $PM_{2.5}$ and NO_X urban increment for all German urban entities was estimated using Equation (1) and the parameters described in Table 1. It should be noted that Equation (1) requires PM_{10} rural background concentration as an input for estimating the urban increment. Considering that measured PM_{10} rural background concentration values are available only for a limited number of urban entities, an alternative approach was used to estimate the values at locations where no data were available. An interpolation of all PM_{10} regional background concentration values available for Germany was considered the best solution for the given conditions. The interpolation method used was the Inverse Distance Weighting (IDW). In this interpolation method, values of a variable are estimated based on the distance from the nearest neighbour of a known point (Davis, 2002). The interpolation method can be written as:

$$z_0 = \frac{\sum_{j=1}^{m} k_j z_j}{\sum_{j=1}^{m} k_j} \quad (3)$$

where z_j are taken from the closest position "*m*" and the weights k_j are chosen larger for data values near to where the estimate z_o is to be made. The essential assumption in this technique is that nearby locations are more likely to have more similar values than when the locations are far apart. In this way, an interpolated geo–referenced grid was generated using the spatial analysis tool included in the software ArcMap.

Once PM_{10} rural background values for all large urban entities were estimated, PM_{10} urban increments were estimated and the modelled average PM_{10} urban increments for fifteen of the over hundred urban entities analysed in this work are depicted as a horizontal line in Figure 3. Results for all cities can be found in the Supporting Material (SM). The vertical bars depict the minimum and maximum urban increment for those urban entities with several rural background concentration values. It should be kept in mind that NO_X rural background concentration is not a parameter to estimate the urban increment, only one value was estimated for each urban entity. The $PM_{2.5}/PM_{10}$ ratio discussed in Section 2.4 was used for estimating the $PM_{2.5}$ urban increment and the results included in the SM. Figure 4 shows the NO_X urban increments for several urban entities.

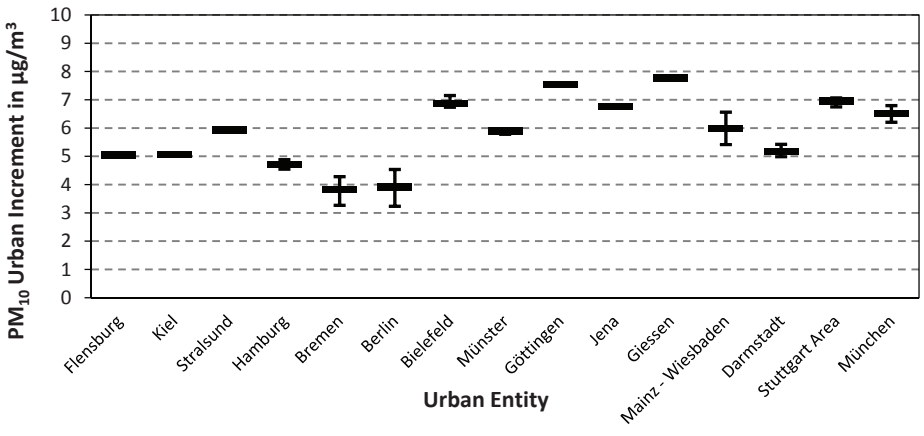

***Figure 3**. PM_{10} urban increment estimated for several large German urban entities.*

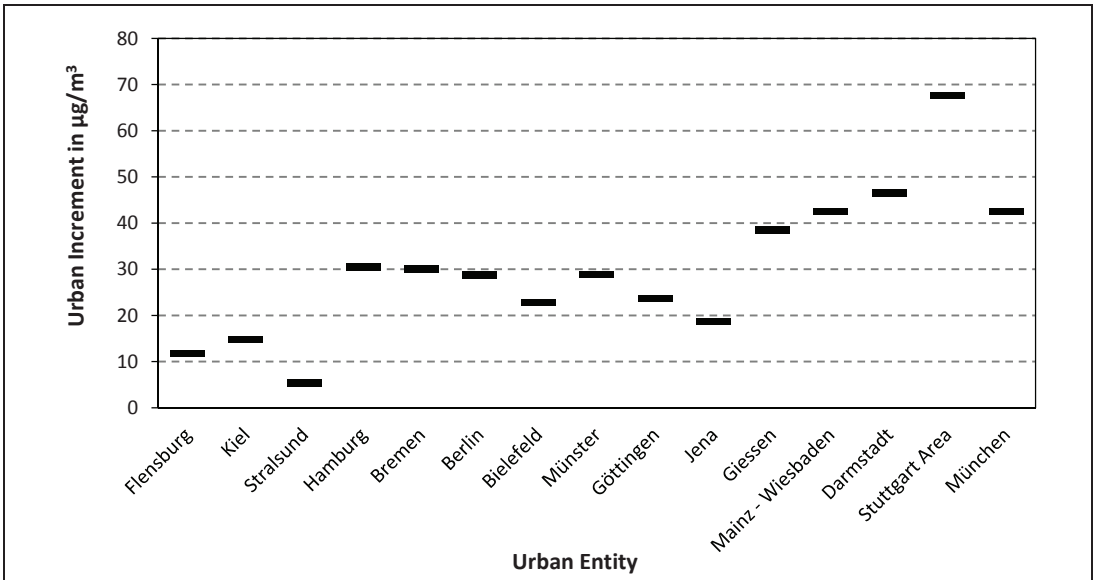

***Figure 4**. NO_X Urban increment estimated for several large German urban entities.*

PM_{10} urban increments for all German urban areas with more than 50 000 inhabitants show values ranging from 2.1 to 10.1 µg/m^3. These values are within the range of measured increments used for developing the model. Furthermore, they are also similar to those reported by Graff (2006). The variability in the urban increment across all cities is likely related mainly to wind speed and emission intensity.

Concerning NO_X urban increments for all German urban areas with more than 50 000 inhabitants, it is pertinent to note that two values over 70 µg/m^3 were obtained. Although differences between urban and rural background concentrations accounting for over 70 µg/m^3 can be found for cities like Milan, Trento and London (Airbase, 2010), such high values are commonly not found in Germany. Moreover, the NO_X model was built using measurements accounting for urban increments from 10 to 58 µg/m^3 and, therefore, urban increments over 70 µg/m^3 are likely to be overestimated (see the SM, Table S1). Finally, it is important to remember that measurements of nitrogen oxides are defined as the sum of nitric oxide and nitrogen dioxide added as a part of per billion and expressed as nitrogen dioxide in µg/m^3 (Mol, 2007). Therefore, despite a detailed spatial disaggregation of NO_X emissions, important chemical processes at the urban scale are difficult to take into account using the simplified approach presented here. Nevertheless, despite minor discrepancies the results presented here support the notion that NO_X concentrations are a significant tracer of the NO_2–equivalent concentrations.

Furthermore, the estimates provide valuable insight into spatial variability of pollutant concentrations across large cities. This information is relevant when evaluating the impact of emission reduction policies on air quality, which should take into account the cost–benefit of each measure. The flexibility for constructing scenarios (i.e., changes in concentration due to emission reductions) is another advantage of the approach. A validation of the computed results by comparison with physical measurements is presented in the next Section.

4. Model Evaluation

In order to implement an air quality modelling system with confidence, it is necessary to evaluate the model performance by means of a comparison of results against measured values. Thus, the performance of the model presented in this work was evaluated by means of comparing the concentration predictions against observations for the reference year 2005. For this purpose, modelled urban increment values were compared against measured urban increment values retrieved from the European Air Quality database Airbase (Airbase, 2010).

The urban increment model described comprehensively in Section 2 was developed using a multiple regression analysis. The predictor variables adopted were: the emissions released within the city; the urban morphology; and the average wind speed. Moreover, the dependent variable – the measured urban increment – was estimated as the difference between observational data from paired rural and urban background measuring stations. From a total of 24 station pairs for PM_{10} and 26 station pairs for NO_X, half of them were used for model development and the other half is used in this Section for evaluating the model. The comparison of the modelled against the measured PM_{10} and NO_X urban increments are shown in Figures 5 and 6. A first visual inspection of the results show an overall good agreement with measurements and the urban increments are basically captured by the dispersion modelling approach. The city of Ingolstadt depicts the highest underestimation for PM_{10} (around 20%) and the highest overestimation was estimated for the city Luneburg (25%). As for NO_X, the city of Ingolstadt depicts the highest underestimation (around 25%) and the highest overestimation was estimated for the city Boblingen (80%).

The model performance was evaluated for its accuracy, bias and errors as well. The metrics applied for this purpose were: the root mean square error (RMSE), the mean fractional bias and error (MFB and MFE, respectively) and the coefficient of determination (R^2). MFB and MFE normalize the bias and error for each model–observed pair by the average of the model and observation before taking the average (Boylan and Russell, 2006). These metrics are presented in Table 2.

***Table 2.** Statistics obtained comparing modelled and measured concentrations at urban background stations for PM_{10} and NO_X*

Pollutant	Measured mean (µg/m^3)	Modelled mean (µg/m^3)	RMSE (µg/m^3)	MFB (%)	MFE (%)	R^2
PM_{10}	6.02	5.75	0.95	5	14	0.71
NO_X	22.9	31.2	12.84	29	34	0.84

RMSE: root mean square error; MFB: Mean fractional Bias; MFE: Mean fractional Error; R^2: Coefficient of determination

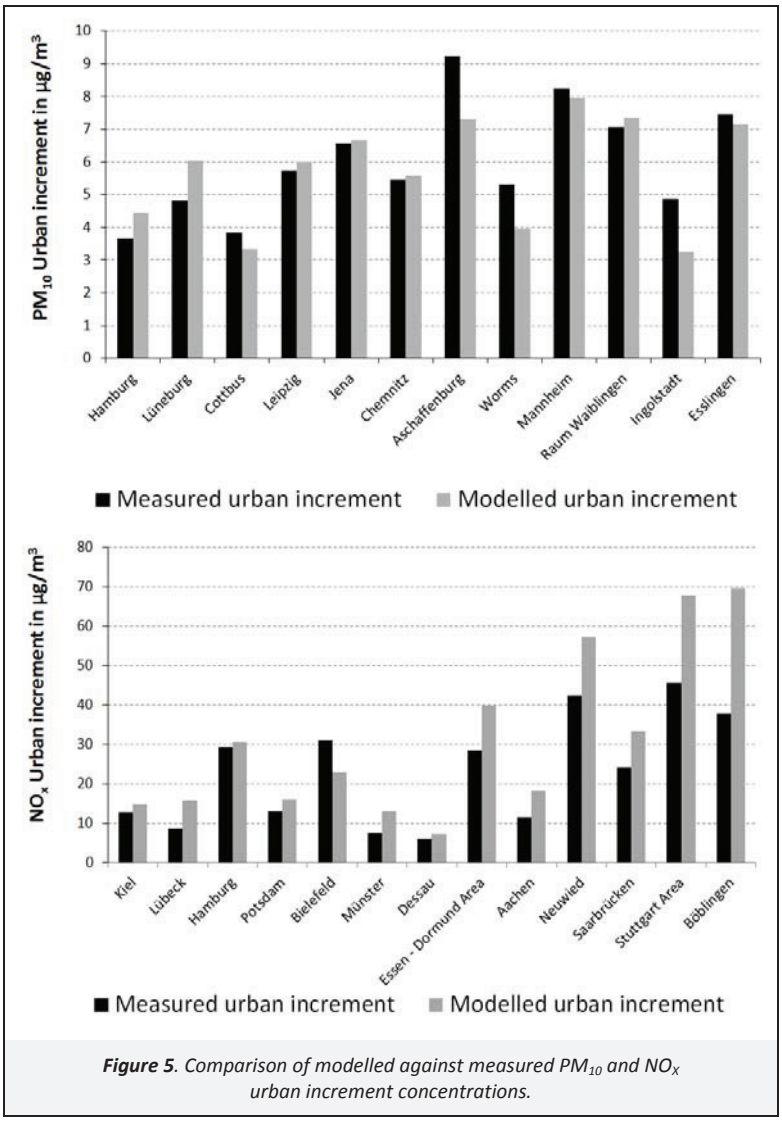

***Figure 5**. Comparison of modelled against measured PM_{10} and NO_X urban increment concentrations.*

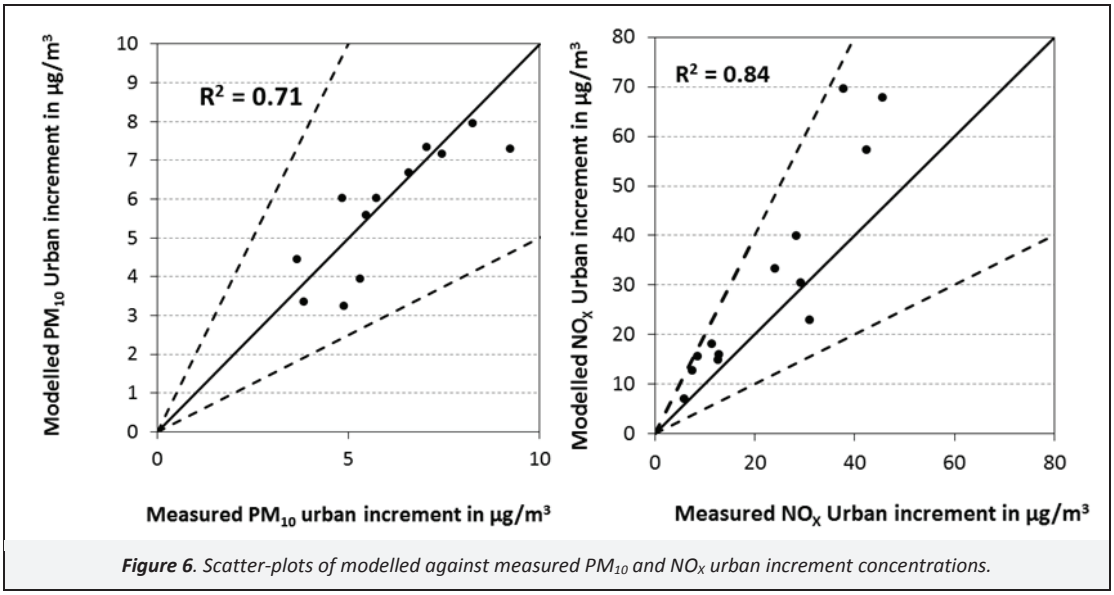

***Figure 6**. Scatter-plots of modelled against measured PM_{10} and NO_X urban increment concentrations.*

The coefficient of determination depicts a strong positive correlation between measured and modelled values for both pollutants. A slightly positive bias in the PM_{10} estimates can be also observed, although the bias for NO_X is clearly larger. Boylan and Russell (2006) suggested, based on results from benchmark studies and examination of various bias and errors metrics, that the model performance goal (defined as the level of accuracy that is considered to be close to the best a model can be expected to

achieve) for particulate matter has been met when both the MFB and MFE are less than or equal to ±30% and +50%, respectively. Furthermore, they also propose that the model performance criteria (defined as the level of accuracy that is considered to be acceptable for standard modelling applications) has been met when both MFB and MFE are less than or equal to ±60% and +70%, respectively. Using these criteria, the model goal performance is met and, although these criteria were developed for particulate matter, the model performance goal is also met for NO_X. Additionally, it is important to note that all modelled values fall within a factor of 2 of the measurements (Figure 6) enforcing the notion that an overall acceptable model performance was accomplished. It is noted as well, that the model performs better for PM_{10} than for NO_X, which may be explained by the higher uncertainty regarding NO_X emissions allocation for urban areas, which is the most relevant parameter influencing the model for urban increment.

In summary, it can be said that the model is useful for the estimation of pollutant concentrations within urban areas. Despite the validation of the modelling approach by means of comparison against measured values, some issues related to model sensitivity and uncertainty remain to be addressed. Due to their inherent importance to the evaluation of any air pollution modelling effort, both aspects are discussed in the SM.

5. Conclusions

In this study, a new model to estimate the ”urban increment”, which is defined as the distinguishable difference between the background concentration in a city and the levels measured outside the urban area, was described. The approach was used for estimating annual concentrations of PM_{10}, $PM_{2.5}$, and NO_2 for all urban areas with more than 50 000 inhabitants in Germany for the reference year 2005. The results showed that there are large differences on air quality levels across the urban areas in Germany, which may be relevant when planning or evaluating emission control strategies. The modelling approach gives an estimation that is as close as possible to the actual concentrations, based on certain quality or goodness criteria. Here it is important to remark that it is possible to use the urban increment model to improve the results of regional models at the urban scale. Two simple approaches for estimating $PM_{2.5}$ and NO_2 concentrations from PM_{10} and NO_X concentrations, respectively, were presented as well. Nevertheless, due to the inherent spatial and temporal variation in the emissions, an application for a more recent year (e.g., 2012) would certainly need a further update of the model’s coefficients.

Moreover, as the modelling approach allows for flexibility of changing initial conditions and building scenarios, the added values of this approach are the large spatial domain covered, the high spatial resolution, and its inherent flexibility to address other environmental issues, such as analysis of emission reduction scenarios, the human exposure to certain pollutants and their associated human health impacts. Further development of the model considers the extension to other EU countries and the inclusion of additional relevant meteorological parameters.

Supporting Material Available

NO_X urban increment for urban entities selected for model building (Figure S1), Discussion on model uncertainty, Diagnostic diagram of the model to estimate PM_{10} urban increment (Figure S2), Diagnostic diagram of the model to estimate NO_X urban increment (Figure S3), PM_{10}, $PM_{2.5}$, NO_X, and NO_2 urban increment for all German urban areas with over 50 000 inhabitants for the year 2005 (Table S1). This information is available free of charge via the Internet at http://www.atmospolres.com.

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